

General Disclaimer

One or more of the Following Statements may affect this Document

- This document has been reproduced from the best copy furnished by the organizational source. It is being released in the interest of making available as much information as possible.
- This document may contain data, which exceeds the sheet parameters. It was furnished in this condition by the organizational source and is the best copy available.
- This document may contain tone-on-tone or color graphs, charts and/or pictures, which have been reproduced in black and white.
- This document is paginated as submitted by the original source.
- Portions of this document are not fully legible due to the historical nature of some of the material. However, it is the best reproduction available from the original submission.

Semi-Annual Progress Report

NASA Grant NGR-14-005-200

"Studies of Basic Mechanisms in High
Pressure Gases - Applications to High Efficiency
High Power Lasers"

Prepared for
National Aeronautics and Space Administration
NASA - Lewis Research Center

1 May 1976

by

J.T. Verdeyen
B.E. Cherrington
S.G. Leslie
W.S. Millar
B.E. Edwards

Gaseous Electronics Laboratory
Department of Electrical Engineering
University of Illinois at Urbana-Champaign
Urbana, Illinois 61801



(NASA-CF-147101) STUDIES OF BASIC
MECHANISMS IN HIGH PRESSURE GASES:
APPLICATIONS TO HIGH EFFICIENCY HIGH POWER
LASEFS Semiannual Progress Report, 1 Nov.
1975 - 1 May 1976 (Illinois Univ.) 23 p HC G3/36

N76-22530
HC \$3.50

Unclass
26797

I. INTRODUCTION

In our recent proposal for contract renewal, we presented preliminary results obtained by optically exciting high pressure Cesium-Xenon mixtures with a high power pulsed dye laser. Since that time, we have concentrated on verifying our initial results through more carefully controlled experiments and extending our measurements to even higher pressures. Our efforts in this direction are discussed in the section immediately following.

The engineering problems encountered when working with the highly reactive alkali metals and dangerously high rare gas pressures has been satisfactorily solved by construction of a stainless steel container. This new cell enhances our ability to work at high Xenon pressures and virtually eliminates the window degradation experienced with the glass containers. The new system, along with the entire experimental apparatus, is described in detail in Section III.

In our renewal proposal, we indicated that other materials show promise as potential laser systems. Section IV reveals some interesting results obtained from our optical absorption measurements made with high pressure Rubidium-Xenon mixtures. In addition, we have achieved a sustained microwave discharge in Rubidium at relatively high Xenon pressures. This is indeed very exciting and is also covered in Section IV.

Finally, we are commencing studies of Cadmium-rare gas mixtures, which are generally considered to be similar to mercury-rare gas systems, in that the lowest excited electronic level is quasi-metastable. Reasons for our interest in Cadmium are briefly mentioned in Section IV.

II. OPTICAL PUMPING OF Cs-Xe MIXTURES

In the recent proposal for contract continuation,¹ it was indicated that the bulk of our investigations regarding Cs-Xe mixtures was directed towards the collisionally induced "forbidden transitions" (7s-6s and 5d-6s). Preliminary results from pulsed dye laser pumping (4600 Å) of the 7p state of cesium were presented and are repeated here for convenience. Figure 1 shows schematically the excited cesium states and the cascade process by which the 7s, 5d and 6p states are populated. Spontaneous fluorescence from the $A^2 \Pi_{3/2}$, $2\Sigma^+$ (formed from the $7^2s_{1/2}$ state), and 2Π (formed from the $5^2d_{3/2}$ state) excimer levels are shown in Fig. 2 for a cesium density of $3.7 \times 10^{17} \text{ cm}^{-3}$ and a Xenon density of $5 \times 10^{19} \text{ cm}^{-3}$ (2 atmospheres at room temperature). It can be seen that the spontaneous emission from the $A^2 \Pi_{3/2}$ level is much larger than emission from either the $2\Sigma^+$ or 2Π states, indicating that a majority of the electrons are cascading down to the 6p atomic states. At higher Xenon densities, however, emission from the 2Σ excimer state overtakes emission from the $A^2 \Pi_{3/2}$ state as shown in Fig. 3. This phenomena suggests that the $2\Sigma^+$ excimer formation rate (which is Xenon pressure dependent in the following manner, $R_f = k_f P^2$) is now dominating the spontaneous decay ($A \sim 7 \times 10^6 \text{ sec}^{-1}$). This is indeed very exciting and it would seem that achieving laser action may be possible. It must be remembered, though, that these data are only preliminary results and that problems were encountered during the experiments.

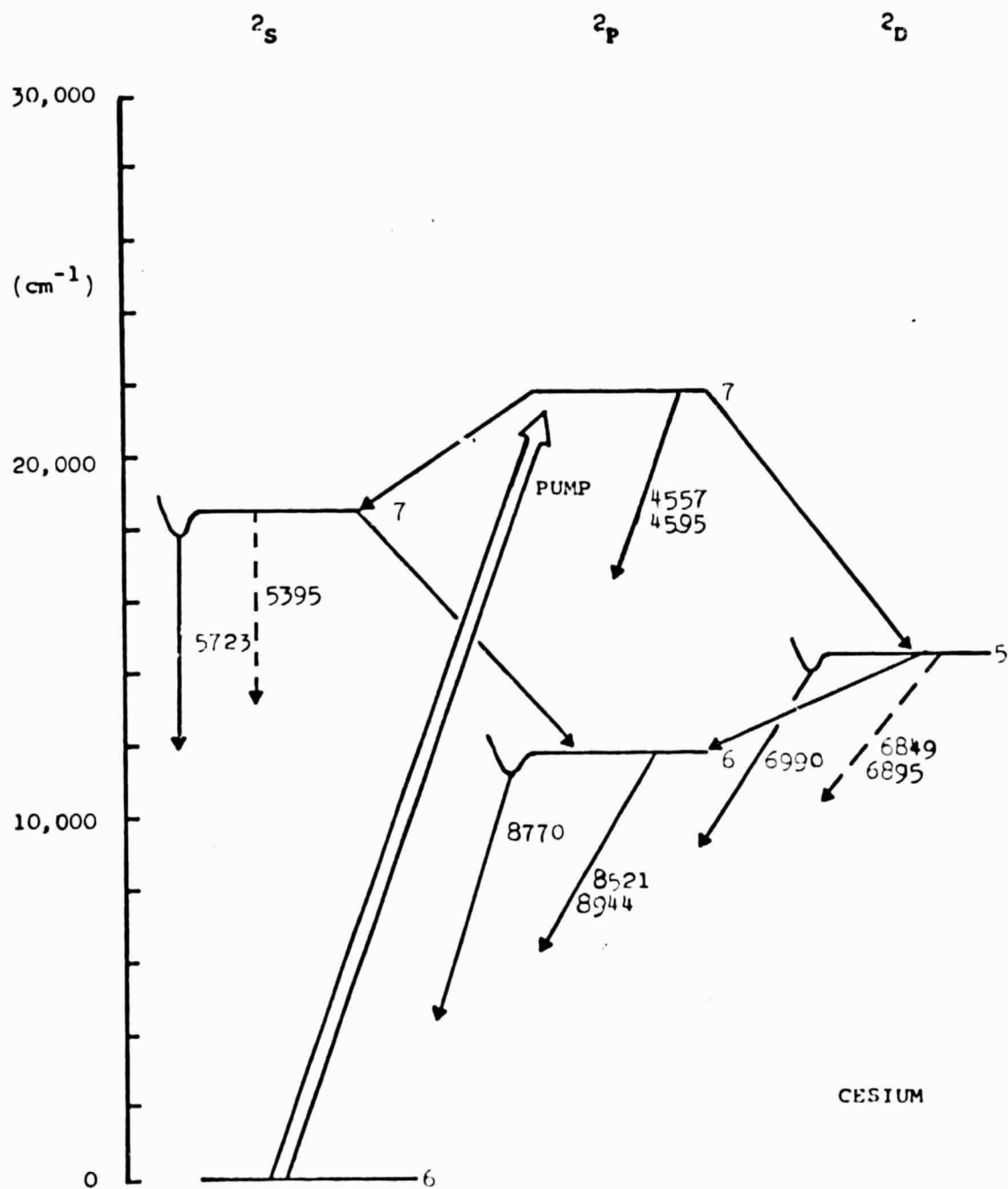


Fig. 1. Partial potential energy level diagram for cesium.

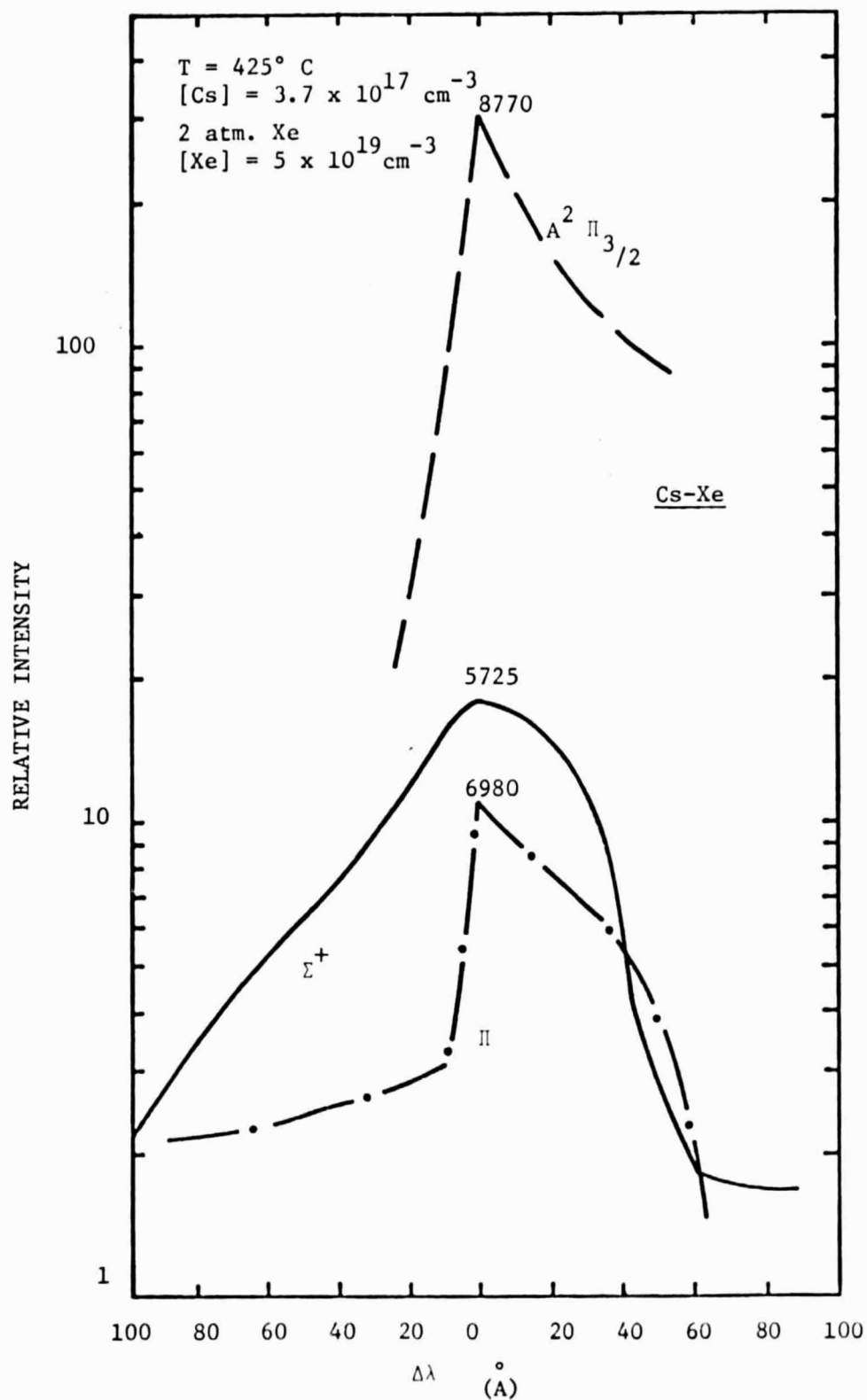


Fig. 2. Relative intensity of the major excimer emissions in dye laser pumped Cs-Xe mixtures for $[\text{Cs}] = 3.7 \times 10^{17} \text{ cm}^{-3}$ and $[\text{Xe}] = 5 \times 10^{19} \text{ cm}^{-3}$.

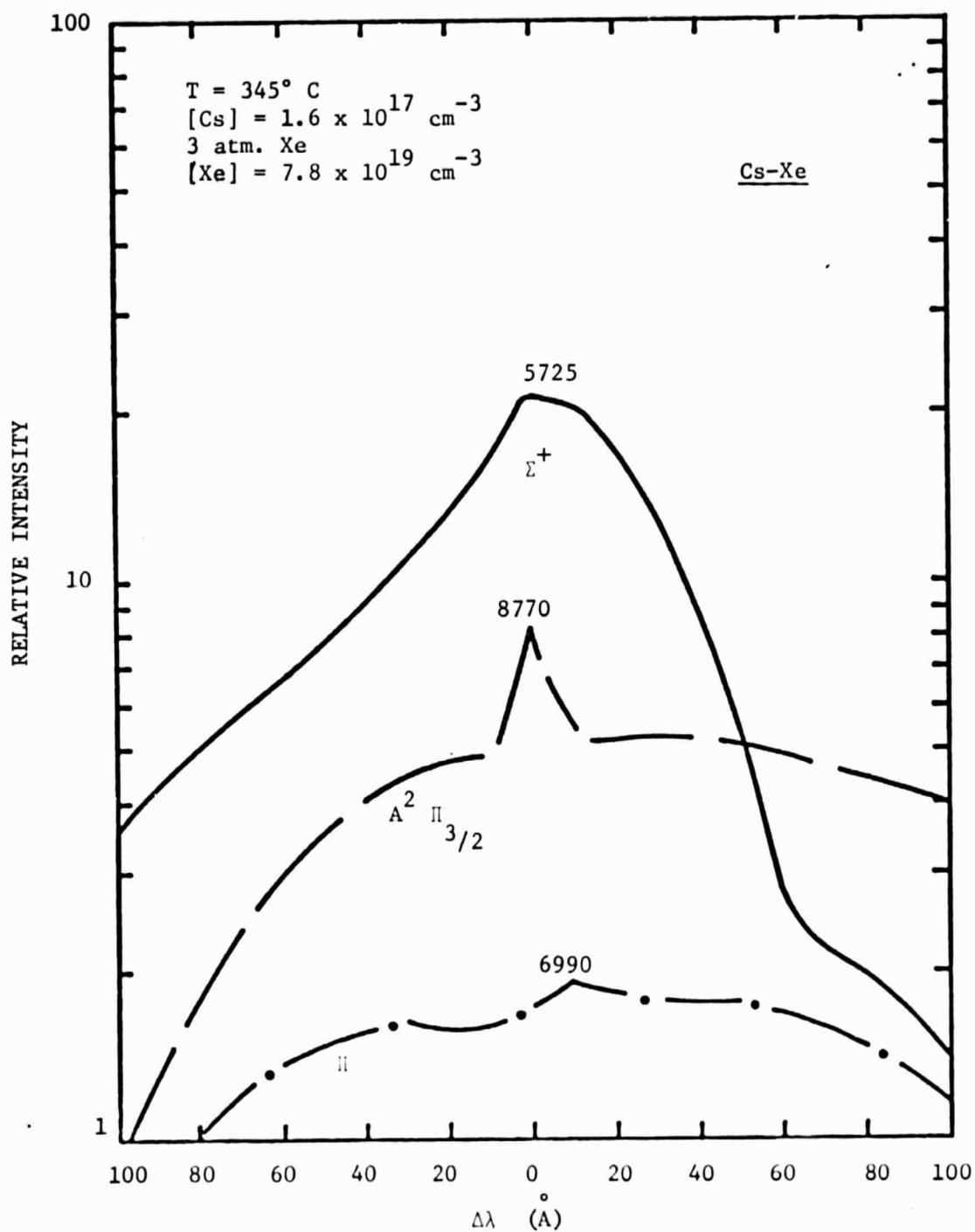


Fig. 3. Relative intensity of the major excimer emissions in dye laser pumped Cs-Xe mixtures for $[\text{Cs}] = 1.6 \times 10^{17} \text{ cm}^{-3}$ and $[\text{Xe}] = 7.8 \times 10^{19} \text{ cm}^{-3}$.

One problem which had a significant effect on the results was the rapid dissociation of the Coumarin-2 dye used in the dye laser. Subsequent diminishing of the laser intensity resulted in the fluorescence weakening as the experiment progressed. To correct for this, the portion of the beam emerging from the "back end" of the laser (totally reflecting mirror) was monitored and the data adjusted accordingly. Since then, it has been shown that this method of monitoring the laser intensity is unsatisfactory. This fact casts a shadow of doubt on the exact results of Fig. 3, but we are certain of the general trends.

A second problem was inherent in the pyrex cells containing the Cs-Xe mixtures. We considered the risk of explosion too great for Xenon pressures above three atmospheres, so we were able to collect data only for Xenon densities below $\sim 8 \times 10^{19} \text{ cm}^{-3}$. Since R_f is pressure dependent, certainly higher pressures would greatly enhance the change over of relative intensities of the Σ^+ and $A^2 \Pi_{3/2}$ levels.

Steps have already been taken to remedy the aforementioned problems. A stainless steel-double windowed container was built to withstand the high pressures required in the experiment. A double window arrangement was used to prevent plating by cesium and is described in detail in the next section. The original container failed, in that the inner sapphire windows were not leak tight, and cesium condensed on the outer windows, which were at room temperature. However, the inner windows are presently mounted in a different fashion and the cesium container is operating quite well.

In order to alleviate the problem of dye laser inconsistency, we decided to use a pulsed argon-ion laser. Because of its consistency and relatively high repetition rates (synchronous detection can be used), the argon-ion laser is a much better choice for diagnostic purposes. To pursue this avenue, an argon-ion laser was constructed, yielding 5 μ sec pulses with an estimated peak power of 3.0 watts.

The 4579 Å line of the argon-ion laser is used to pump both the red wing of the broadened cesium $7^2 p_{3/2}$ state (line center = 4557 Å) and the blue wing of the $7^2 p_{1/2}$ state (line center = 4594 Å). Here, the general plan is to use the argon-ion laser to reaffirm the data presented in Figs. 2 and 3 and to gain insight into the Cs-Xe system tendencies at higher rare gas pressures. When a favorable condition for laser action is found, the argon-ion laser will then be replaced by the pulsed dye laser as an optical pump source.

At present, optical absorption measurements are being taken to determine the cesium density at which the spontaneous fluorescence intensity is a maximum. This is not trivial in that the pump line is not at line center and the atomic resonance line must be broadened enough to obtain a sufficient amount of absorption of the pump intensity. A sample absorption run previously done with a 25 cm long pyrex cell filled with 3 atmospheres of Xenon ($7.8 \times 10^{19} \text{ cm}^{-3}$) at $\sim 230^\circ \text{ C}$ ($[\text{Cs}] = 10^{16} \text{ cm}^{-3}$) is shown in Fig. 4. According to the absorption profile shown in Fig. 4, the line broadening is more than sufficient to overlap the 4579 argon-ion laser line. In view of this knowledge, operation of the cell at temperatures higher than 200° C should result in sufficient absorption of the pump intensity.

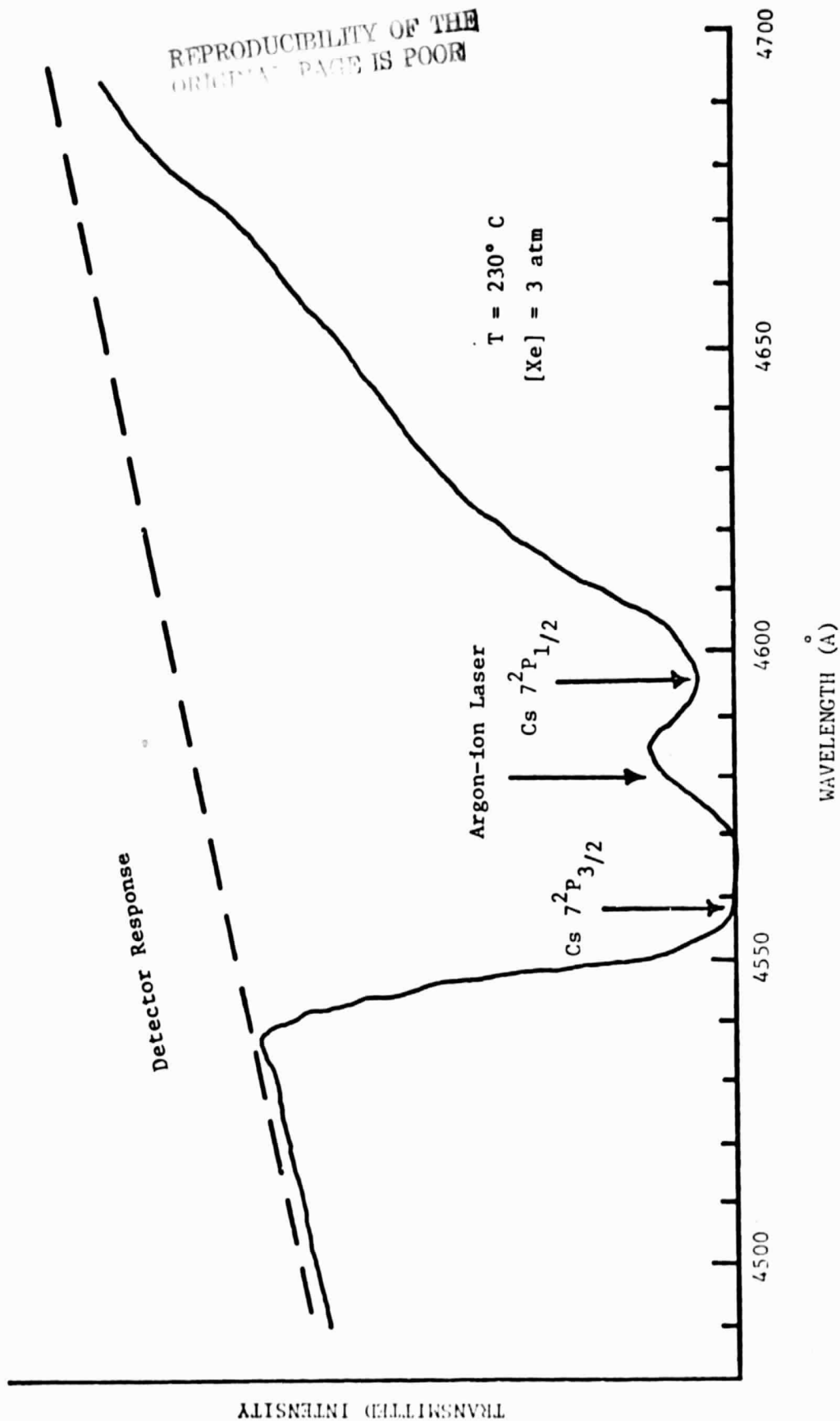
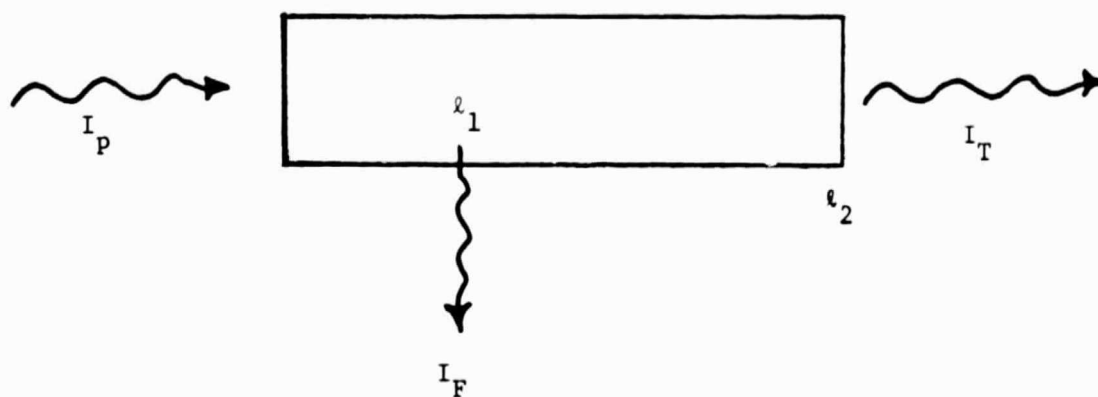


Fig. 4. Absorption profile of the cesium 7^2P states as a function of wavelength for $[\text{Cs}] = 10^{16} \text{ cm}^{-3}$ and $[\text{Xe}] = 7.8 \times 10^{19} \text{ cm}^{-3}$.

Due to the fact that the Cs-Xe cell is not optically thin and the fluorescence is being collected a finite distance away from the optical pump input, an upper limit is imposed on the temperature at which the cell can be operated. The optimum temperature at which to operate the cell can be determined by the following simple reasoning. Consider a cell where the fluorescence is being collected at ℓ_1 at right angles to the axis of the cylinder.



If the absorption coefficient of the Cs-Xe mixture at the 4579 Å argon-ion line is α , we can write an expression for the fluorescent intensity I_F as follows:

$$I_F = K I_A = K \alpha I_P e^{-\alpha \ell_1},$$

where I_A is the absorbed intensity and K is some proportionality constant. To find the maximum fluorescent intensity, we take the

derivative of I_F with respect to α and set it equal to zero,

$$\frac{dI_F}{d\alpha} = K I_p e^{-\alpha \ell_2} - K \alpha \ell_1 I_p e^{-\alpha \ell_1} = 0,$$

and we find that the stipulation $\alpha \ell_1 = 1$ must be satisfied in order to obtain a maximum I_F .

In order to experimentally determine the conditions under which the above condition occurs, absorption measurements must be made with the following thought in mind. To begin, the transmitted intensity can be found by the following method:

$$I_T = I_p e^{-\alpha \ell_2}.$$

However, it has already been determined that $\alpha \ell_1 = 1$, so α can be written as $\frac{1}{\ell_1}$. This leaves us with the following expression for I_T :

$$I_T = I_p e^{-\frac{\ell_2}{\ell_1}}.$$

In other words, the fraction of transmitted light (I_T/I_p) at 4579 Å must equal $e^{-\ell_2/\ell_1}$ in order to obtain the maximum fluorescence at ℓ_1 . For the Cs-Xe cell currently in use, this fraction $\sim .02$ or 2% transmission.

Absorption measurements are currently being made to determine the cell temperature at which the above condition is satisfied. As soon as a viable temperature range is found, the argon-ion laser will be used to pump the Cs-Xe mixture and the resulting behavior of the fluorescence with Xe pressure investigated.

In summary, because of difficulties in taking data utilizing a pulsed dye laser with a slow repetition rate, an argon-ion laser was constructed. With the argon-ion laser, pump power is sacrificed but we are able to work at higher repetition rates and thus use synchronous detection techniques. In addition, a stainless steel cell was built to contain high pressure (> 5 atmospheres) cesium-xenon mixtures. At present, absorption measurements are being taken to find the conditions under which the fluorescence from the cesium excited states is a maximum. As soon as these measurements are terminated, the argon-ion laser will be used to study the fluorescence due to the "forbidden transitions" from the $2P^+$, 2Π and $A^2\Pi_{3/2}$ states. Once favorable conditions for laser action are found, the high power pulsed dye laser will be used to optically pump the mixture.

ORIGINAL FROM FILE

III. EXPERIMENTAL APPARATUS

The recent contract continuation proposal¹ indicated that the major obstacle to continued study of Cesium-Xenon excimer formation was the mechanical limitations of the sealed glass tube configuration. In addition to the dubious ability of a glass cell to withstand high Xenon pressures, it was noted that the windows of the glass cell undergo rapid degradation due to continuous contact with hot Cesium vapor. Another serious problem was the combined effect of the saturated vapor pressure of Cesium and temperature gradients in the sealed tube to form a "fog" of Cesium metal droplets, which further deteriorated the system's optical quality.

In order to solve these problems, the construction of a stainless steel cell with thickwall windows protected by sapphire windows and a temperature controlled reservoir to control Cesium vapor pressure was suggested as a solution. At present, this newly designed system is complete and operational.

The stainless steel cell is illustrated in Fig. 5. The cell consists of three sets of dual windows (the weaker inner windows being of Cesium resistant sapphire and the thicker outer windows of Pyrex) contained in stainless steel Varian nipples. The inner windows are mounted in mechanically tight fittings, which limit the outward flow of Cesium vapor. Necessarily, these fittings permit a slow equalization of Xenon pressure between the hot inner region, which contains the Cesium vapor, and the cooler outer region, which is Cesium-free. Cooling coils, mounted externally at the extremities of the cell, preserve the integrity of the temperature sensitive

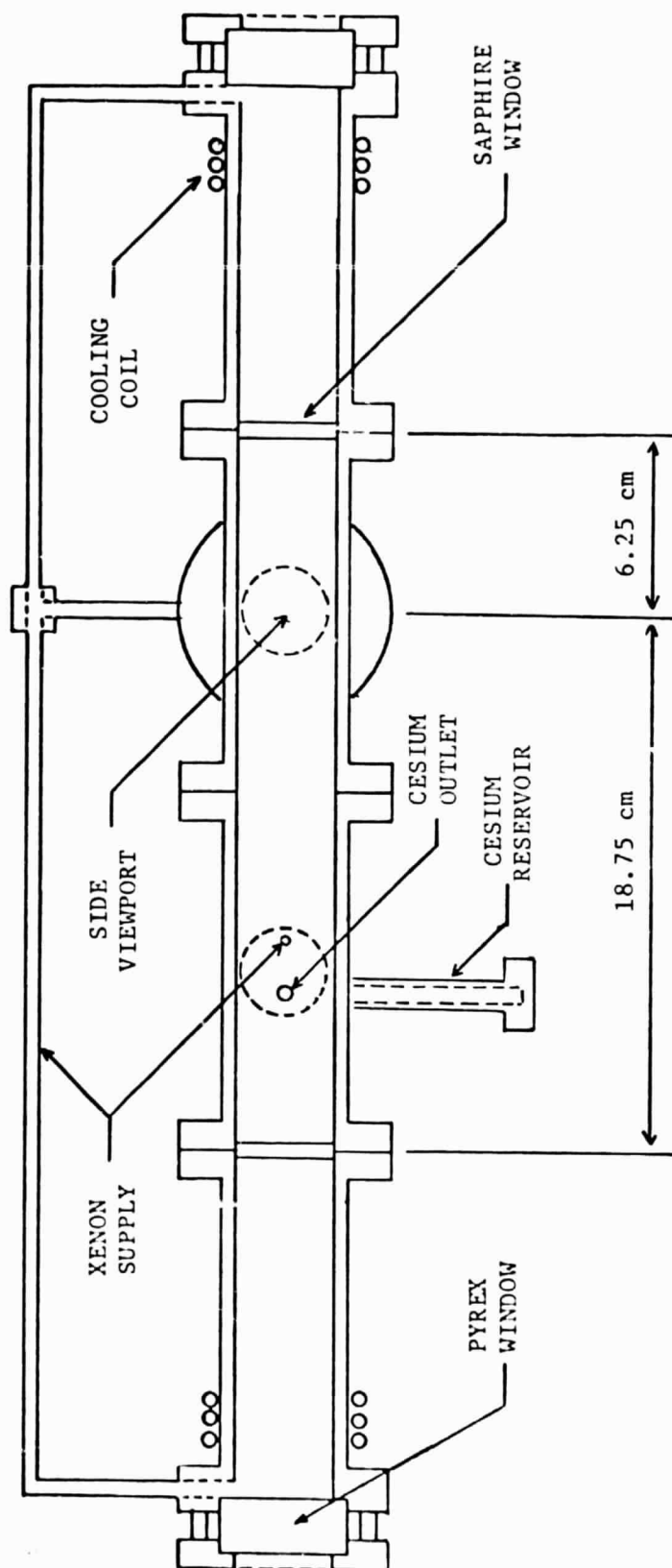


Fig. 5. Schematic diagram of the stainless steel cell used to contain high pressure Cs-Xe mixtures.

rubber O-rings, which seal the thick Pyrex windows to the system. Cesium vapor enters the inner region of the cell through stainless steel tubing from a physically separate Cesium reservoir.

The high pressure cell is surrounded by an oven, which maintains the desired temperature of the excimer formation region. A separate oven controls the temperature of the Cesium reservoir. This provides a means of adjusting the Cesium vapor pressure in the main cell.

Initially, both regions of the stainless steel cell are filled with a predetermined amount of Xenon gas. At this point, the system is sealed from the external Xenon supply line, and slowly heated to its operating temperature. This temperature is calculated to yield the appropriate Xenon density in the excimer formation region after pressure equalization between the hot and cool regions of the cell has occurred. Finally, Cesium vapor is added to the cell by heating the reservoir to a temperature slightly below that of the main oven.

A schematic diagram of the entire experimental apparatus is shown in Fig. 6. A pulsed Argon ion laser optically excites the Cs-Xe mixture. Consistent excitation of the mixture is assured by continuously monitoring the shape and power of the output pulse. Fluorescence from the mixture is sampled through a side viewport in the cell and focused onto the slits of a scanning monochromator. The amplitude and wavelength of the optical emission are automatically recorded on a strip chart. Also included in the schematic is a dye laser, which will be used in the future to optically pump the Cs-Xe mixture in hopes of inducing laser action in the Cs-Xe cell.

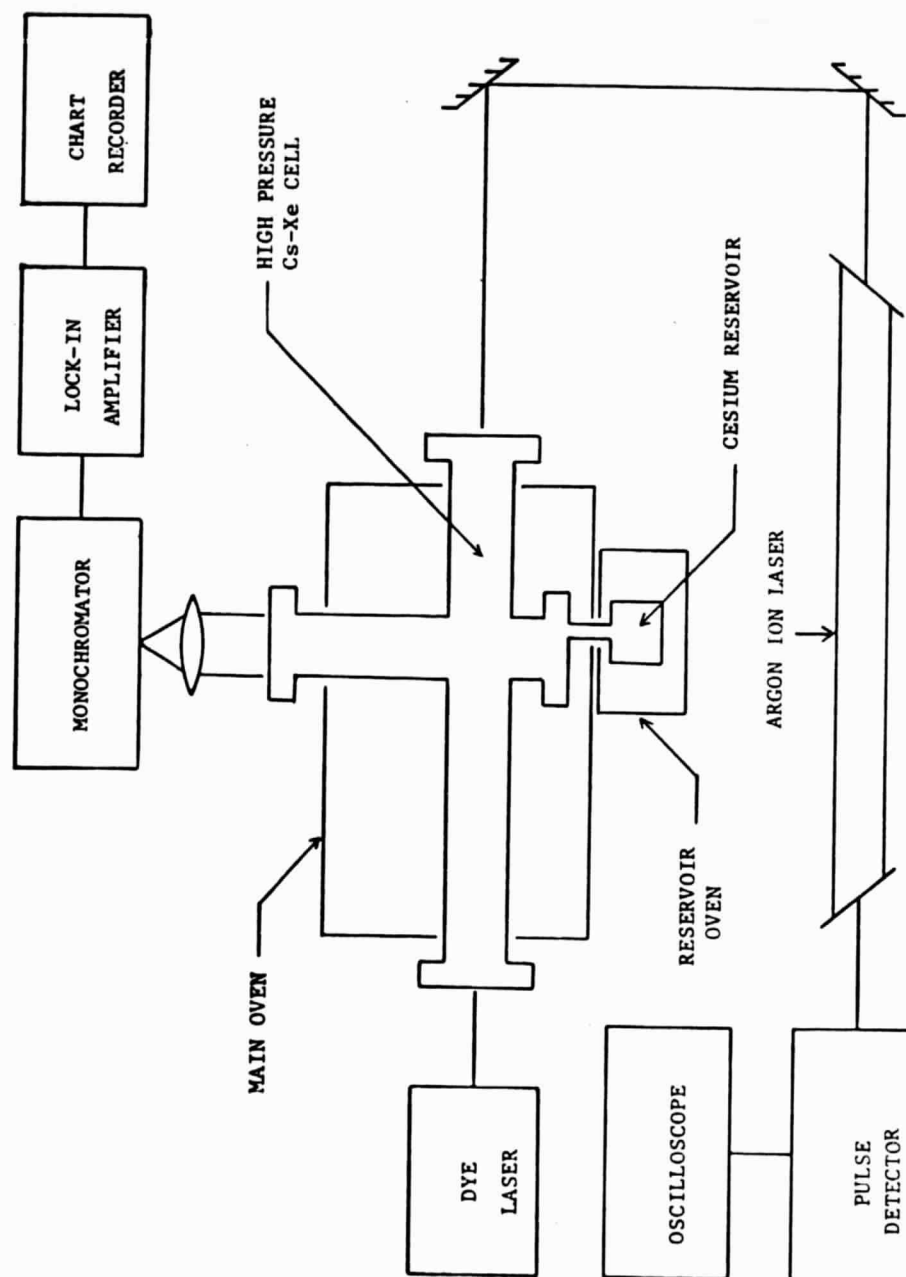


Fig. 6. Experimental apparatus used for optical excitation experiments with high pressure Cs-Xe mixtures.

IV. SPECTROSCOPY OF OTHER "FORBIDDEN" TRANSITIONS

In addition to our work in Cesium-Xenon, we have begun to study other metal vapors to see if their "forbidden" transitions will also yield an excimer.

Rubidium was the natural choice for our first experiment, since it is almost identical to Cesium in energy level structure. Consider Fig. 7 which is an energy level diagram of Rubidium. Based upon our experience with Cesium, we would expect excimer bands to the red of the 4967 \AA and 5167 \AA "forbidden" transitions. The bottom graph of Fig. 8 represents one of our absorption experiments. While this curve clearly shows "forbidden" transitions occurring, the band structure is rather complicated. The $4^2D \rightarrow 5^2S$ transition seems to exhibit the normal "forbidden" line, followed by the excimer red wing. However, explaining the two absorption peaks at 5016 \AA and 5090 \AA is not as easy. The 4^2D and 6^2S levels are almost degenerate, and their molecular potential curves rather involved, giving rise to non-curve crossing effects.⁽²⁾ A reasonable explanation may be that the 5016 \AA absorption is associated with the $6^2S \rightarrow 5^2S$ transition, while the 5090 \AA absorption is a blue shift due to the $4D \Sigma$ potential, the shape of which is quite different from the other $4D$ potential curves.

In addition to absorption work, we have just completed apparatus for the study of Rb-Xe spectra from a microwave sustained discharge. Figure 9 is a schematic diagram of this equipment, consisting of a 30 watt CW magnetron coupled into a resonant cavity.

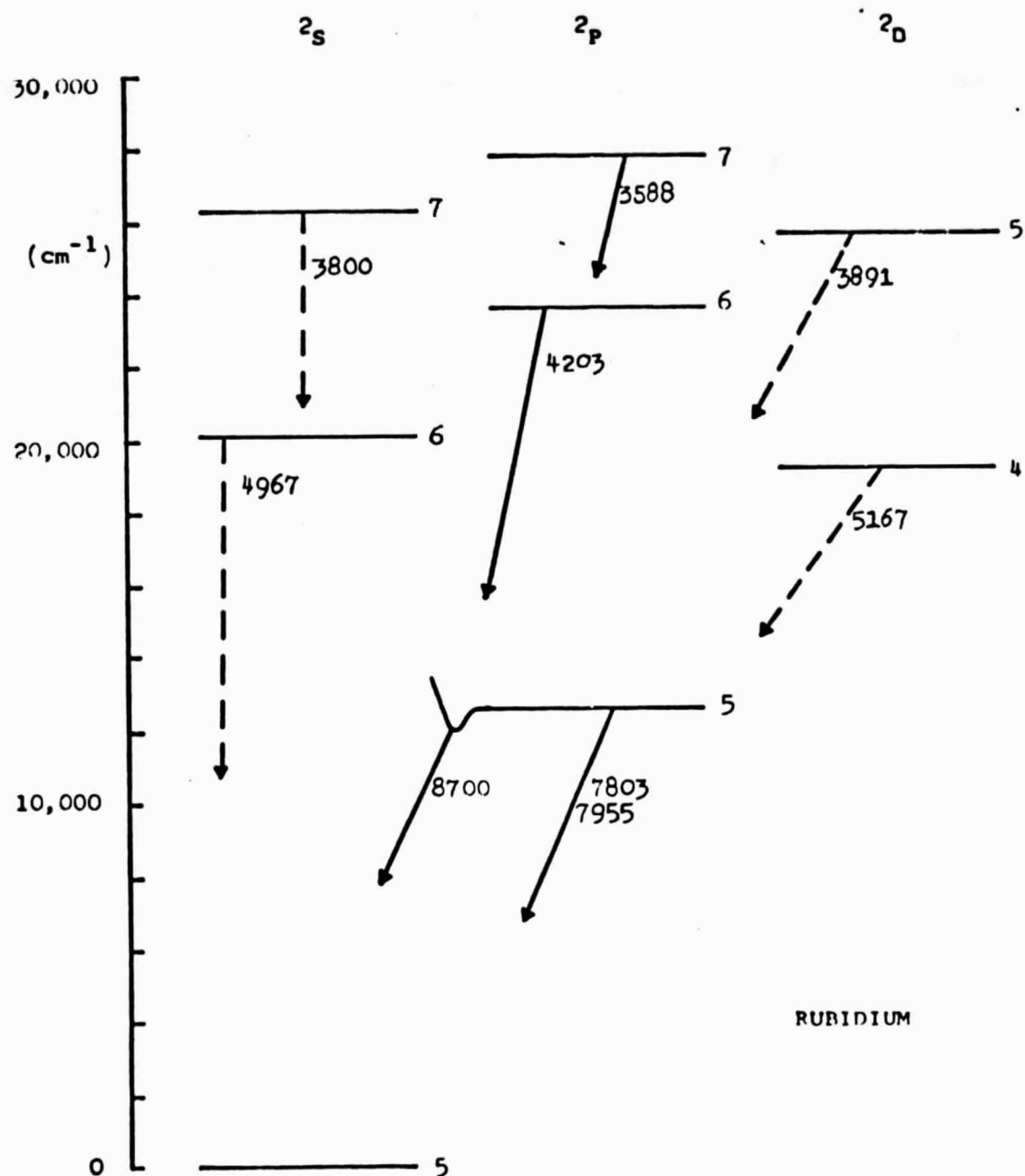


Fig. 7. Partial energy level diagram of atomic Rubidium along with the expected transition due to Rb Xe excimers. Forbidden transitions are shown by dashed lines.

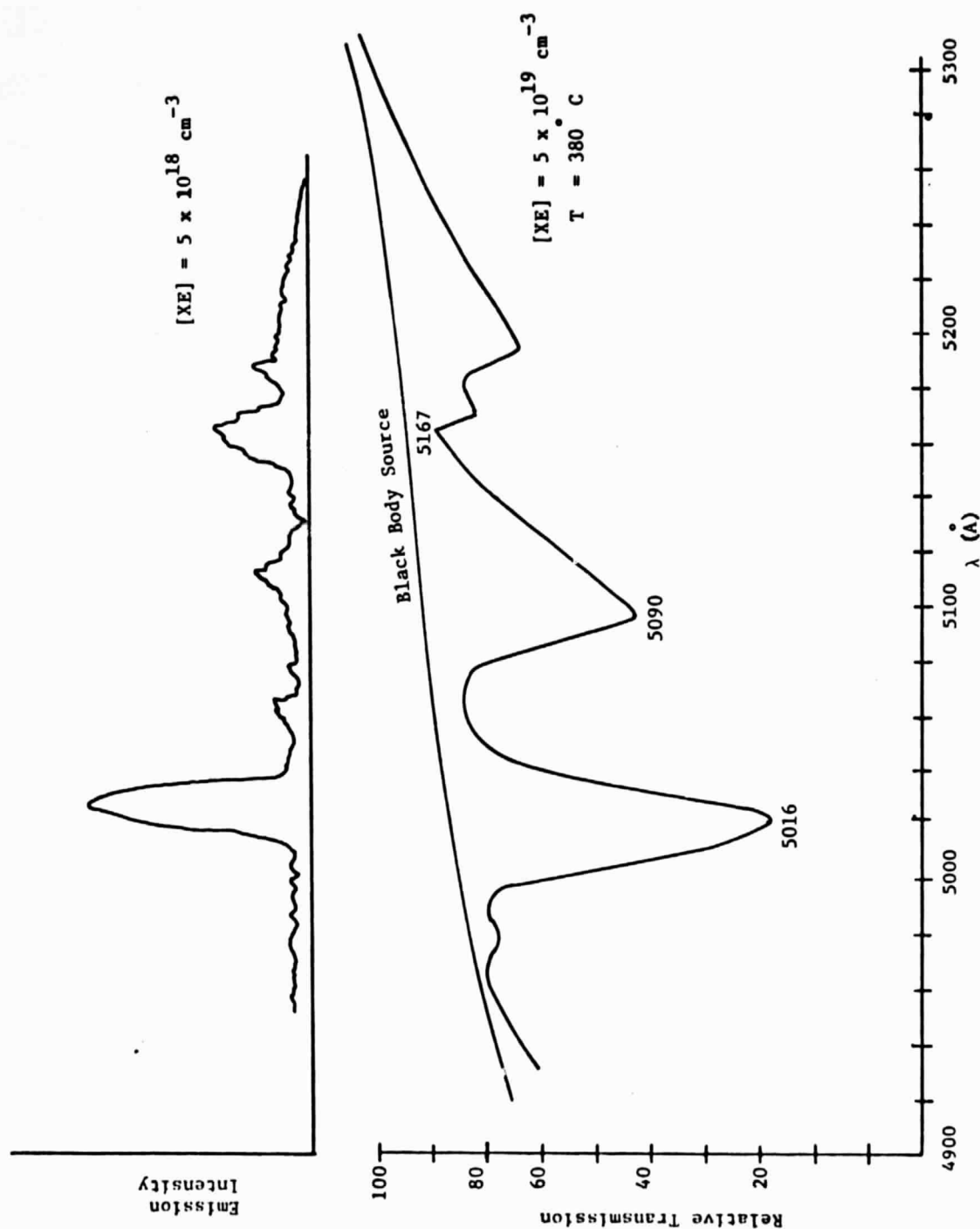


Figure 8. Absorption and Emission Spectra of Rb Xe

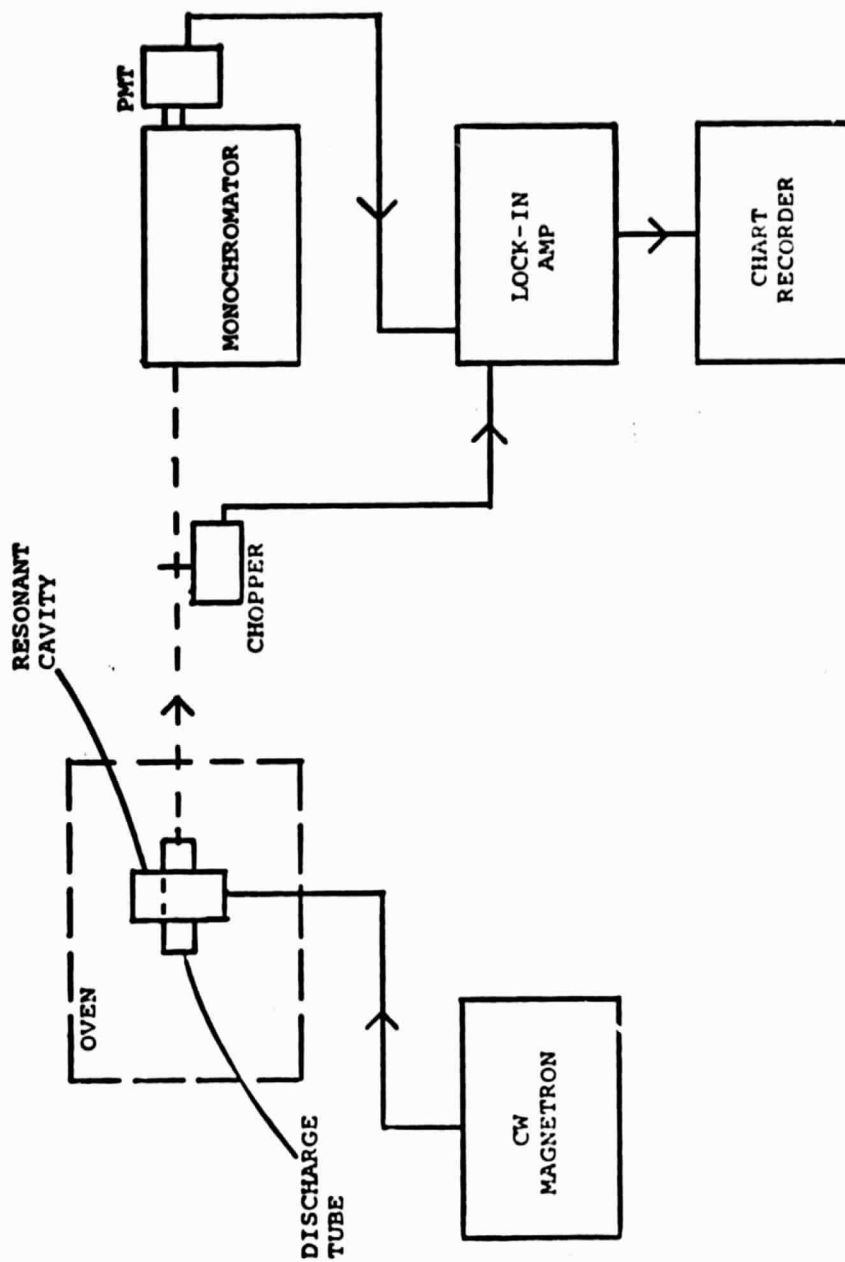


Fig. 9. Microwave apparatus used to achieve a sustained discharge in Rb-Xe.

The cavity sits inside an oven to maintain Rb density. Early results indicate that this approach will be quite successful. We have easily maintained a glow discharge in 650 torr of rare gas, and foresee higher pressures in the near future. The upper graph of Fig.8 is the emission spectrum from a Rb-Xe mixture at 200 torr. It represents only preliminary data based on the first discharge that we took a spectrum from.

Up until now, we have concentrated on the alkali metal - rare gas systems. However, there are some other metal vapor - rare gas systems which are also potentially attractive. Cadmium is one such system. Being isoelectronic with mercury, Cadmium's lowest excited state is "quasi-metastable," with a very weak "forbidden" transition occurring from $^3P_1 \rightarrow ^1S$, similar to the 2537 \AA transition in Hg, giving it the possibility of high energy storage. If an excimer can be formed from one of the 3P states, this large energy storage capability could be put to use as a laser. Whether or not the excimer actually forms is what we intend to find out.

Evaluation of the Cadmium - rare gas system is just now reaching the experimental stage. We will be studying this system in both absorption and emission to see if its "forbidden" transition configuration may be of possible use.

BIBLIOGRAPHY

- [1] Verdeyen, J.T., Cherrington, B.E., Leslie, S.G., Millar, W.S.,
and Edwards, B.E., "Studies of Basic Mechanisms in High Pressure
Gases - Applications to High Efficiency High Power Lasers,"
Proposal for the Continuation of NASA Research Grant
NGR-14-005-200, December, 1975.
- [2] Pascale, J., and Vandeplanque, J., J. Chem. Phys. 60, 2278 (1974).